IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

Shigeru YAO, et al. Group Art Unit: 1771

Serial No.: 09/539,929 Examiner: Hai Vo

Filed: March 31, 2000

For: POROUS INSULATING FILM AND ITS LAMINATES

DECLARATION UNDER 37 C.F.R. 1.132

Assistant Commissioner of Patents and Trademarks,
Washington, D.C. 20231

Sir:

I, Yoshihiro KUSUKI, c/o Polymer Research Laboratory, Ube Industries, Ltd., 8-1, Goiminamikaigan, Ichihara-shi, Chiba, Japan do hereby declare:

That I am a Docter of Technology and a research chemist by profession having been employed by Ube Industries, Ltd., Yamaguchi, Japan.

That I have been engaged in research work in the area of gas separation membrane, polymer synthesis and polymer physics, and provided many research reports and obtained many patent rights as shown in Lists A and B

attached hereto.

That I have read the specification of the above-identified U.S. application (hereinafter referred to as "present application" for brevity) and hence I am fully familiar with the invention disclosed in the present application;

That I have read and am fully familiar with the art cited against the claims of the present application;

That, to confirm the results described in Example 4 of U.S. Patent No. 5,510,395 to Tomioka et al. (hereinafter referred to as Tomioka), I carried out the following experimental working.

Experimental Working

The following results are described in Example 4 of Tomioka.

Hydrogen gas permeability: 7.0×10^0 $cm^3/cm^2 \cdot sec \cdot cmHg$, which corresponds to a resistance to passage of hydrogen gas of 2.1 sec/100 cc according to JIS-P-8117

Separation ratio: 2.2 which can be seen from the separation ratio (hydrogen permeability/nitrogen

permeability) curve in Fig. 1 of Tomioka

In order to confirm the above results, a porous film was prepared, by repeating the procedures described in Example 4 of Tomioka, as follows. That is to say, 8.0009g of diaminodiphenylether was dissolved in a mixed solvent of 119.44g of THF and 31.82g of methanol at 13°C, and the resulting sollution was kept at 13°C. Then, 8.8064g of pyromellitic dianhydride was added to the solution at a time, and the mixture was continued to be stirred for 18 hours at 20°C. Thus, a uniform yellow solution as described in Example 4 of Tomioka was obtained. The obtained polyamic acid had an intrinsic viscosity of 1.32. To this solution, 29.79g of water was added to prepare a uniform film-forming solution.

This film-forming solution was cast onto a glass substrate at a speed of about 40 mm/s using a doctor knife with a slit of 500 μm. After drying at 25°C for 20 minutes, the thus formed film was separated from the glass substrate and heated at 80°C for 2 hours and then at 300°C for 3 hours to effect imidization. In this way, a porous film as described in Example 4 of Tomioka was obtained.

The observation of a sectional SEM image of the film showed the presence of a large number of elliptic pores as

described in Example 4 of Tomioka. The elliptic pores were found to exist independently one by one, as is seen from the sectional SEM images of Figs. 1 and 2 attached hereto.

Further, the observation of a surface SEM image of the film proved an uneven surface having no open pores, as is seen from the surface SEM images of Figs. 3 and 4 attached hereto in which Fig. 3 shows the surface opposit to the glass substrate side surface and Fig. 4 shows the glass substrate side surface.

The film has a density, a thickness, a hydrogen gas permeability, a nitrogen gas permeability and a separation ratio (i.e., hydrogen gas permeability/nitrogen gas permeability ratio) as shown below.

Density: 0.61 g/cm^3

Hydrogen gas permeability: 6.45×10^{-4} $cm^3/cm^2 \cdot sec \cdot cmHg$, which corresponds to a resistance to passage of hydrogen gas of 23,000 sec/100 cc according to JIS-P-8117

Nitrogen gas permeability: 2.42×10^{-4} $cm^3/cm^2 \cdot sec \cdot cmHg$, which corresponds to a resistance to passage of hydrogen gas of 62,000 sec/100 cc according to JIS-P-8117

Separation ratio: 2.7

The resistance to passage of air could not be measured as being more than 7,200 sec/100 cc.

Further, the above-mentioned experiment was repeated at a 1/2 sacle and the similar results were obtained.

I, the undersigned declarant, declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and; further, that these statements were made with knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001, of Title 18, of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 1st day of September, 2003

Yoshihiro Kusuki

Yoshihiro Kusuki

List A

K. Okamoto, K. Tanaka, H. Kita, A. Nakamura, Y. Kusuki	J. Polymer Sci., Polym. Phys Ed., 27, 1221 (1989).	The effect of Morphology on Sorption and transport of carbon dioxide in a polyimide from 3,3',4,4'-biphenyltetracarboxylic dianhydride and 4,4'-oxidianiline.
K. Okamoto, K. Tanaka, T. Shigematu, H. Kita, A. Nakamura, Y. Kusuki	Polymer, 31(4), 673 (1990).	Sorption and Transport of carbon dioxide in a polyimide from 3,3',4,4'-biphenyltetracarboxylic dianhydride and dimethyl-3,7-diaminodibenzothiophene-5,5-dioxide.
K. Tanaka, H. Kita, K. Okamoto, A. Nakamura, Y. Kusuki	Polymer journal, 22(5), 381 (1990).	Gas Permeability and Permselectivity in Homo- and Copolyimides from 3,3',4,4'-Biphenyltetracarboxylic Dianhydride and 3,3'- and 4,4'-Diaminodiphenylsulfones.
N. Tanihara, K. Tanaka, H. Kita, K. Okamoto, A. Nakamura, Y. Kusuki, K. Nakagawa	J. Chem. Eng. Japan, 25(4), 389 (1992).	Vapor-permeation separation of water-ethanol mixtures by asymmetric polyimide hollow fiber membrane modules.
Y. Hirayama, T. Yoshinaga, Y. Kusuki, K. Ninomiya, T. Sakakibara, T. Tamari	J. Membrane Sci., 111 (1996), 169.	Relation of gas permeability with Structure of Aromatic Polyimides I.
Y. Hirayama, T. Yoshinaga, N. Kusuki, K. Ninomiya, T. Sakakibara, T. Tamari	J. Membrane Sci., 111 (1996), 183.	Relation of gas permeability with Structure of Aromatic Polyimides II.
Y. Kusuki, H. Simazaki, N. Tanihara, S. Nakanisi, T. Yoshinaga	J. Membrane Sci., 134 (1997) 245.	Gas permiation properties and characterization of asymmetric carbon membranes prepared by pyrolyzing asymmetric polyimide hollow fibre
H. Suzuki, K. Tanaka, H. Kita, K. Okamoto, H. Hosino, T. Yoshinaga, Y. Kusuki,	J. Membrane Sci., 146 (1998) 245.	Preparation of composite hollow fiber membranes of poly(ethylene oxide)-containing polyimide and their CO2/N2 separation properties.
K. Okamoto, S. Kawamura, M. Yoshino, H. Kita, Y. Hirayama, N. Tanihara, Y. Kusuki,	Ind. Eng. Chem. Res., 38 (11), 4425 (1999).	Olefin/Paraffin Separation through Sarbonized Membranes Drived from an Asymmetric Polyimide Hollow Fiber Membrane

Y. Hirayama, Y. Kase, N. Tanihara, Y. Sumiyama, Y. Kusuki, K. Haraya	J. Membrane Sci., 160 (1999) 87.	Permeation properties to 002 and N2 of poly(ethylene oxide)-containing and crosslinked Polymer films.
Y. Hirayama, N. Tanihara, Y. Kusuki, Y. Kase, K. Haraya, K. Okamoto	J. Membrane Sci., 163 (1999) 373.	Permeation properties to hydrocarbons, perfluorocarbons and chlorofluorocarbons of cross-linked membranes of polymethacrylates with poly(ethylene oxide) and perfluorononyl moieties.
Y. Hirayama, T. Yoshinaga, S. Nakanishi, Y. Kusuki,	ACS Symposium Series 733, Polymer Membranes for Gas and Vapor Separation, 1999, p-194, ACS, Washington, DC	Relation between Gas permeabilities and Stracture of Polyimides.
S. Morooka, K. Kusakabe, Y. Kusuki, N. Tanihara	Membrane Science and Technology Series, 6, Resent advances in gas separation by Microporous by ceramic membranes, 2000, p-323, Elsevier	Microporous carbon membranes.

List B

United States Patent No. 5,064,446

United States Patent No. 5,141,642

United States Patent No. 5,286,539

United States Patent No. 5,744,575

United States Patent No. 6,210,464

United States Patent No. 6,464,755

Fig.3

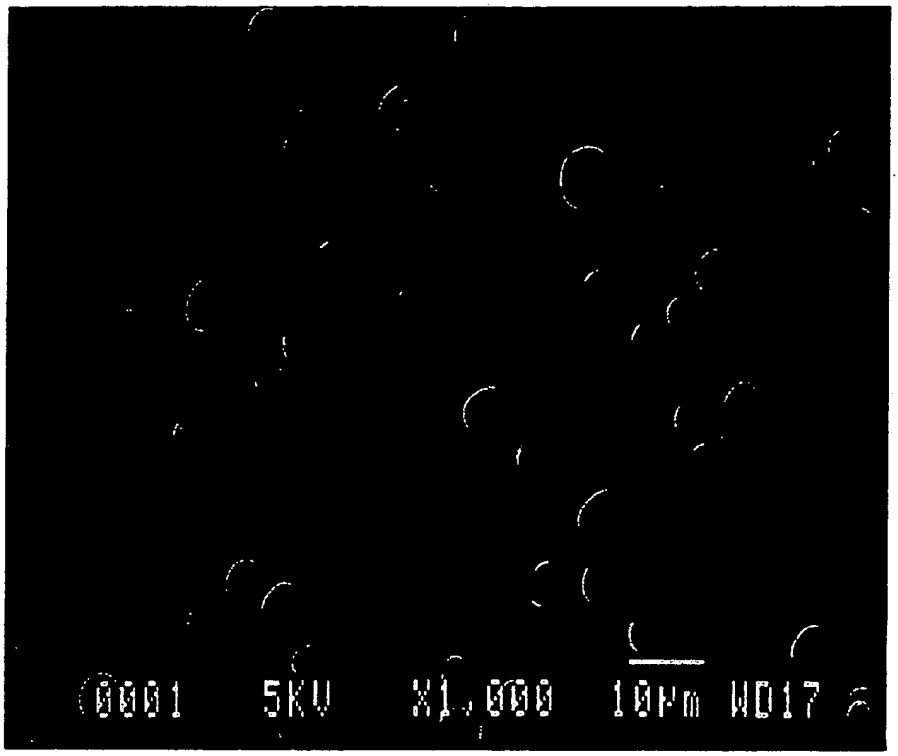

Fig.4

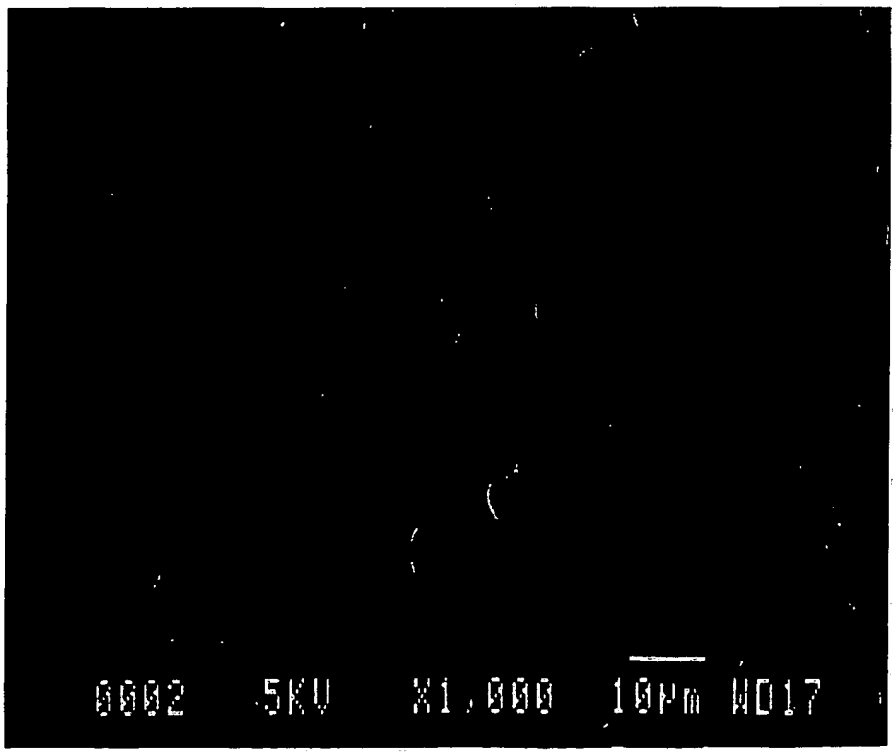

Fig.1

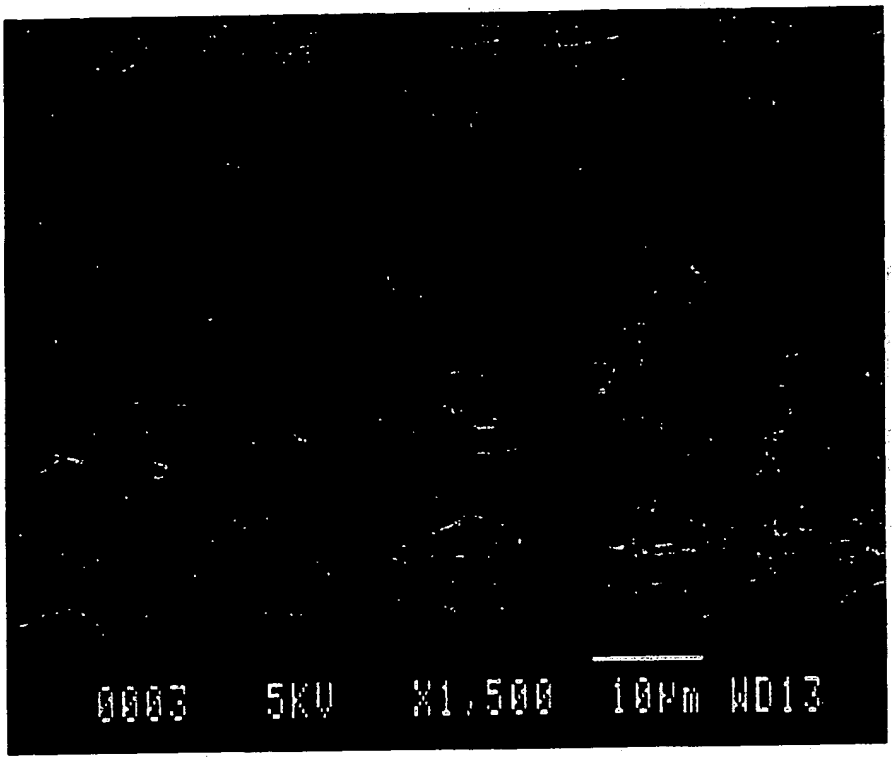

Fig.2